Declaration of Dr. Choon K CHAI

I, Choon K CHAI, am currently employed by INEOS Olefins & Polymers Europe in the position of senior Polymer Specialist in Rheology and Processing at the Research and Technology Centre in Brussels, Belgium since August 2004.

From July 1995 to August 2004, I was employed by BP Chemicals in the position of Senior Polymer Scientist and Team Leader, in charge of the Rheology Laboratory at the Research and Technology Centre, Lavéra, France. I was also BP Chemicals Rheology Skill Network Leader in coordinating and networking materials sciences skills across company research sites in USA, UK and France.

Previous to this I was employed as Senior Polymer Scientist, Polyolefins Catalysis Research and Development. In this position I had the responsibility for the materials sciences in Polyolefins catalysis research at Sunbury, England. I had been in that position since September 1993.

Prior to this, I was Senior Polymer Scientist, Physical Sciences Branch at BP Chemicals' Sunbury Research Centre, in England, where I was responsible for researching new processing development for BP Solar thin-film technology from 1988 to 1990. I held various research positions at Sunbury from the time of my entry to BP in September 1980. These positions included Research Physicist (1980 - 1986), senior Research Physicist and Deputy Project Leader (1986 - 1993) in polymer sciences research in the Materials Sciences Branch at Sunbury.

Before joining BP, I worked as a Research Scientist in the Polymer Physics Division at the Natural Rubber Producers Research Association (MRPRA) in Brickendenbury, Hertford (September 1972 - September 1977). I received my PhD in Polymer Engineering and Physics from the University of Oxford in England in 1980, and graduated from London University with First Class Honours in Physics with Mathematics in 1977.

I am the sole inventor of the present application (Serial Number 10/567,927) and have read and understood the cited references US 2003/0215659 (Farley) and US 2004/0053022 (Ohlsson).

A copolymer of ethylene and 1-hexene was prepared according to the procedure described in the examples of the present application. The copolymer had a density of 0.912 g/mol, a melt index (g/10ml) of 20 measured under a load of 2.16 kg at 190°C, a melt elastic modulus G' (G"=500Pa) of 16 Pa measured at 190°C, and a molecular weight distribution (Mw/Mn) of 2.8.

The copolymer is therefore representative of Example 3 of the present invention and will be referred to as Copolymer A for the purposes of this declaration.

Copolymer A was analysed with respect to the key polymer properties described in both Farley and Ohlsson.

US 2003/0215659 (Farley)

Farley teaches blends of (a) very low density polyethylenes (VLDPE's) and (b) low density polyethylenes (LDPE) which may be used for extrusion coating applications. Farley describes the VLDPEs as being prepared by means of bisCp metallocenes rather than monoCp metallocenes and in particular unbridged bisCp zirconocenes such as bis(1,3-methyl,n-butyl cyclopentadienyl) zirconium dichloride.

Table IV of Farley discloses a number of polymers including inventive sample A made with such metallocene catalysts. Although there is little data, apart from density and melt index, with respect to the copolymers described in Farley the key parameters of Compositional Distribution Breadth Index (CDBI), melt index ratio and Temperature Rising Elution Fractionation (TREF) are all indicative of the molecular structure, thus produced by such metallocene catalysts, that in turn influences the rheology of the copolymers.

The broad disclosure of Farley indicates copolymers having a CDBI in the range 55 – 70% with exemplification in Table IV for sample A of

64.5%. The melt flow ratios disclosed are in the range 6 – 15 and the polymers exhibit two peaks in TREF.

Copolymer A representative of the copolymers suitable for use as component (a) of the present invention has been analysed for the above properties of CDBI, TREF and melt index ratio.

CDBI is defined to be the percent of polymer whose composition is within 50% of the median comonomer composition. It is calculated from the composition distribution curve and the normalized cumulative integral. The CDBI of linear polyethylene, which does not contain a comonomer, is defined to be 100%. The CDBI of Copolymer A was determined using a procedure similar to that described in Farley and well established in other patent applications to ExxonMobil the assignee of Farley. The CDBI for Copolymer A was found to be 86%.

The TREF curve for Copolymer A was obtained using an analytical scale method and clearly shows a single peak as follows:
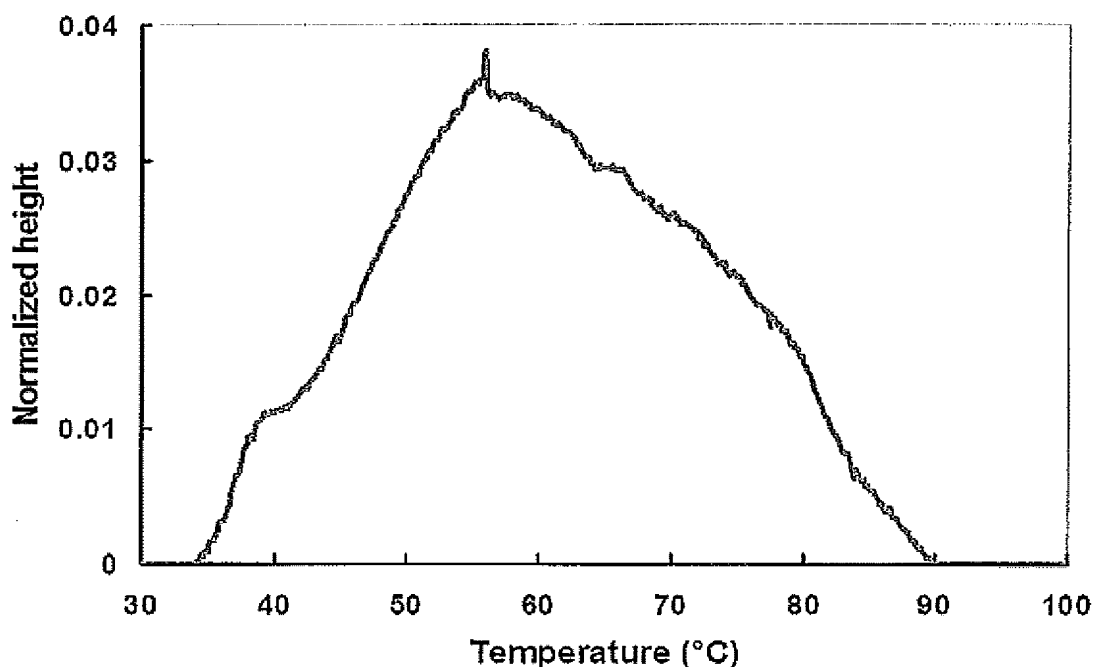
The melt index ratio is defined as the ratio I_{21}/I_2, where I_{21} is the melt index measured under a load of 21.6 kg, and I_2 is that under 2.16 kg, at 190°C. It was found to be impossible to measure the melt index of Copolymer A under 21.6 kg as it was too fluid under these conditions but it was possible to determine the melt index (I_{10}) under a load of 10 kg and hence to determine the melt index ratio of I_{10}/I_2.

The melt index value under a load of 10 kg was $I_{10} \sim 140$, giving the I_{10}/I_2 ratio:

$$I_{10}/I_2 = 140/20 = 7$$

However, one can estimate the I_{21}/I_2 from I_{10}/I_2 with the following relationship obtained from the proportionality in the log-log plot of the melt index value against the corresponding applied load:

$$[\log(I_{10}/I_2)]/[\log(I_{21}/I_2)] = [\log(10/2.16)]/[\log(21.6/2.16)]$$

that is, their ratios are related to the log of their loads ratios (i.e., 10 kg against 2.16 kg; 21.6 kg vs. 2.16 kg), giving the equation

$$I_{21}/I_2 = (I_{10}/I_2)^{1.5}.$$

Thus, for Copolymer A having $I_{10}/I_2 = 7$ as shown above, its estimated $I_{21}/I_2 = 18.5$

To summarise the data as follows:

Parameter	Copolymer A	General disclosure (Farley)	Sample 2 (Farley)
CDBI	86%	60 – 80%, preferably 55 – 70%	64.5%
TREF	1 peak	2 peaks	
I_{21}/I_2	18.5	6 – 15, preferably 9 - 12	17.2

Farley describes metallocene-produced VLDPEs which are preferably linear polymers ie, without long chain branching. With respect to the metallocenes used for their preparation these are preferably bisCp métallocènes, and in particular are preferred catalyst systems which will minimize or eliminate long chain branching, rather than monoCp metallocenes used in the present application.

Based on their methods of preparation it would be expected that the copolymers described in Farley would exhibit different rheological properties to those of the copolymers of the present invention in particular with respect to the degree of long chain branching. This is clearly evidenced from the above analysis of CDBI, TREF and melt index ratio that elucidate the significant differences in molecular structure between the copolymers of the present invention and those described in Farley.
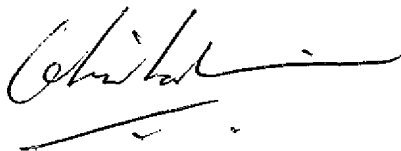
US 2004/0053022 (Ohlsson)

The copolymers of Ohlsson exhibit a melt index ratio (I_{21}/I_2) in the range 30 to 80 and from Table 2 resin examples 1 and 2 exhibit ratios of 46 and 57.6 respectively. From the above analysis of Copolymer A of the present invention the melt index ratio is found to be 18.2 thus well outside the range of the copolymers of Ohlsson and hence strongly indicative as to the significant differences in molecular structure between the copolymers of Ohlsson and those of the present invention.

Conclusions

Based on the measured properties of Copolymer A which is representative of component A of the present invention and the description, examples and reported data in both Farley and Ohlsson I consider that the copolymers described in either reference are different to those claimed in the present invention.

Dr C Chai
Brussels

27th May 2010